

# Achieving Superplasticity in Fine-Grained Al-Mg-Sc Alloys

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**Abstract.** Superplasticity denotes the ability of a limited number of materials to achieve exceptionally high tensile elongations of at least 400%. Experiments show that the Al-Mg-Sc alloys provide excellent capabilities for achieving superplastic flow and also they can be formed easily in biaxial superplastic forming operations. It is important, therefore, to examine the superplastic flow mechanism when the alloy is prepared using different procedures. This report examines the superplastic characteristics of these alloys after preparation without subjecting to any severe plastic deformation (SPD), after processing using the two SPD procedures of equal-channel angular pressing (ECAP) and high-pressure torsion (HPT) and after processing using the alternative procedure of friction stir processing (FSP). The results are compared using each technique and they are examined with reference to a theoretical model that was developed specifically for superplastic flow in conventional alloys.

## Introduction

The mechanical properties of metals may be evaluated very easily by pulling samples in tension and measuring the flow stress and the tensile elongation to failure. Generally, when metals are pulled in tension, they break after only relatively small amounts of elongation. In some situations, however, it is possible for the metal to pull out to a very high elongation in the process known as *superplasticity*. In formal terms, the process of superplasticity is now defined specifically as an elongation to failure of at least 400% and an associated strain rate sensitivity close to  $\sim 0.5$  [1]. From an historical perspective, there were reports in early experiments of quite large tensile elongations to failure, such as 163% in brass in 1912 [2] and  $\sim 300\%$  in Cd-Zn and Pb-Sn alloys in 1928 [3] but these elongations are not sufficient to qualify as superplastic behavior. Accordingly, the first report of true superplasticity was in 1934 when experiments by Pearson in England showed that it was possible to achieve a tensile elongation of  $\sim 1950\%$  in a near-eutectic Bi-Sn alloy [4]. Although this result was innovative and remarkable, it was overlooked by the western scientific establishment but it was pursued extensively by several investigators in the Soviet Union [5]. Ultimately, a comprehensive review of the Russian work was published in English in a western journal [6] and this led to a series of experiments on superplasticity that were initiated at MIT [7]. Following this early work, experiments on superplastic flow spread to many laboratories around the world so that it is now an established research activity in the field of Materials Science. Many of the early results were described in a review celebrating the seventy-five years since the advent of superplastic flow [1].

A first consideration suggested that superplasticity was probably only a scientific curiosity but later it became apparent that the use of superplastic metals in biaxial forming operations provided a potential for achieving remarkably smooth curved structures that were not easily attained using conventional metallurgical processing. As a result, the superplastic forming industry developed rapidly to the extent that it now processes thousands of tons of metallic sheet metals each year for use in a wide range of applications including in the aerospace and automotive sectors and for architectural embellishments [8,9]. It is important, therefore, to investigate the process scientifically and to identify the factors that are critical in allowing metallic samples to exhibit superplastic elongations.

There are two fundamental requirements for achieving superplastic flow [10]. First, there must be a very small grain size, typically smaller than  $\sim 10 \mu\text{m}$ . Second, since superplasticity is a diffusion-controlled process, the tensile testing or forming operation must be conducted at a high temperature, typically at or above  $\sim 0.5T_m$  where  $T_m$  is the absolute melting temperature. In general, these two requirements are incompatible because small grains tend to grow at elevated temperatures in pure metals or simple solid solution alloys. This means that superplastic metals are generally two-phase eutectic or eutectoid alloys or they may contain a fine dispersion of a second phase to act as a grain refiner.

### The nature of the flow process in superplasticity

When materials pull out in superplastic flow, very high elongations are attained but the grains within the sample remain reasonably equiaxed. This suggests that the flow process is grain boundary sliding (GBS) in which the grains slide over each other but they retain approximately their overall shapes. A detailed analysis of flow in superplasticity suggests that GBS accounts for all of the strain incurred in superplastic deformation [11]. Nevertheless, if flow occurs in this way then there must be some limited intragranular dislocation slip in order to accommodate the flow and to prevent the build up of large cracks or cavities within the sample. Several experimental results are now available confirming the occurrence of limited dislocation slip during the flow process but where this slip makes no net contribution to the total strain [12-15].

It was shown in an early analysis that superplasticity may be achieved in samples when the grain size is smaller than the subgrain size [16]. Thus, subgrains are formed within the larger coarse grains during conventional high temperature deformation where these subgrains have boundaries with low angles of misorientation. However, analysis showed that in superplastic materials the equilibrium subgrain size,  $\lambda$ , is larger than the grain size,  $d$ , so that no subgrain boundaries are formed within the grains [16]. Using this approach, Fig. 1 shows the accommodation of GBS in (a) a conventional coarse-grained material where  $d > \lambda$  so that subgrains are present and (b) a superplastic material where  $d < \lambda$  and the individual grains are very small. In practice, GBS leads to stress concentrations at triple points as at A and C in Fig. 1 and this concentration produces an accommodation by dislocation slip in the next grain with the dislocations flowing across the grain and climbing to the subgrain boundaries as at B or to grain boundaries as at D. In this model, the superplastic strain rate is given by a relationship of the form [17]:

$$\dot{\epsilon}_{\text{SP}} = \frac{AD_{\text{gb}}G\mathbf{b}}{kT} \left(\frac{\mathbf{b}}{d}\right)^2 \left(\frac{\sigma}{G}\right)^2 \quad (1)$$

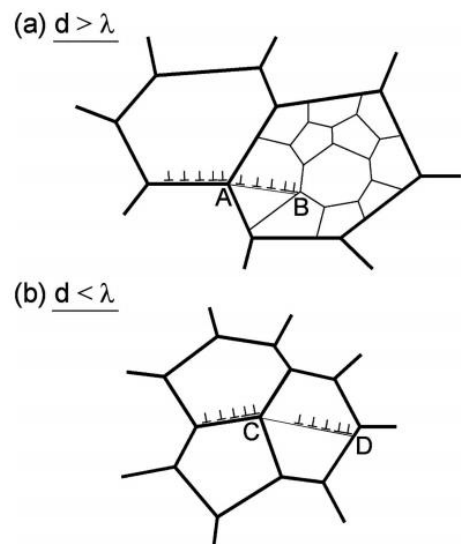


Fig. 1 Accommodation of GBS by slip in (a) a coarse-grained material and (b) a superplastic material [17].

where  $D_{gb}$  is the coefficient for grain boundary diffusion,  $G$  is the shear modulus,  $\mathbf{b}$  is the Burgers vector,  $k$  is Boltzmann's constant,  $T$  is the absolute temperature,  $\sigma$  is the flow stress, the exponents of the inverse grain size and the stress are both equal to 2 and  $A$  is a dimensionless constant with a value of  $\sim 10$ . A detailed analysis shows that Eq. (1) is in good agreement with the superplastic behavior in a range of Al and Mg alloys [18].

### The characteristics of thermal stability in Al-based alloys

In order to achieve superplasticity in aluminium-based alloys, it is necessary that a very small grain size is retained at a high testing temperature within the range associated with diffusion-controlled processes. Figure 2 illustrates the difficulty of achieving this objective where the various datum points represent different samples and each sample was held at the annealing temperature for one hour and then measurements were taken to determine the average grain size. All samples were processed by equal-channel angular pressing (ECAP) which is a procedure for processing by severe plastic deformation (SPD) where the sample undergoes no significant change in dimensions during the processing operation and these samples were pressed through totals of 4, 6 or 8 passes (p) [19]. Thus, it is apparent that pure Al is not stable at elevated temperatures since the grains grow rapidly at temperatures above  $\sim 500$  K. This lack of thermal stability means that it was not possible to achieve true superplastic elongations in samples of pure Al processed by ECAP through 4 passes at room temperature and then tested in tension at a temperature of 403 K [20]. Furthermore, the Al-3% Mg solid solution alloy exhibited a much small grain size after ECAP processing but again there was rapid grain growth at temperatures above  $\sim 500$  K.

By contrast, the Al-3% Mg alloys containing additions of Zr or Sc showed not only very small grain sizes after processing by ECAP but also, for the alloys having 0.2% Sc or 0.2% Sc plus 0.12% Zr, there was reasonable thermal stability and grain sizes  $< 1.0 \mu\text{m}$  up to at least  $\sim 700$  K. This suggests that it may be feasible to achieve good superplastic elongations in an Al-3% Mg alloy with an addition of 0.2% Sc.

A very clear example of true superplastic behavior is shown in Fig. 3 where samples of the Al-3% Mg-0.2% Sc alloy are shown after pulling to failure under different processing conditions. All samples were pulled in tension at 523 K using an initial strain rate of  $1.0 \times 10^{-3} \text{ s}^{-1}$  and the samples were processed prior to tensile testing using ECAP for 8 or 10 passes (p) or by high-pressure torsion (HPT) for 10 turns (t). The lack of necking within the gauge length for the sample showing an elongation of 1020% matches the requirements for superplastic flow [22].

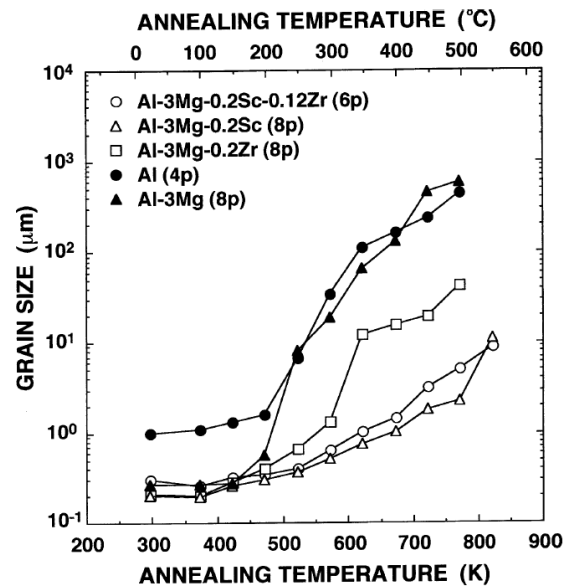


Fig. 2 Grain size versus annealing temperature for samples annealed for one hour after ECAP through 4, 6 or 8 passes (p) [19].

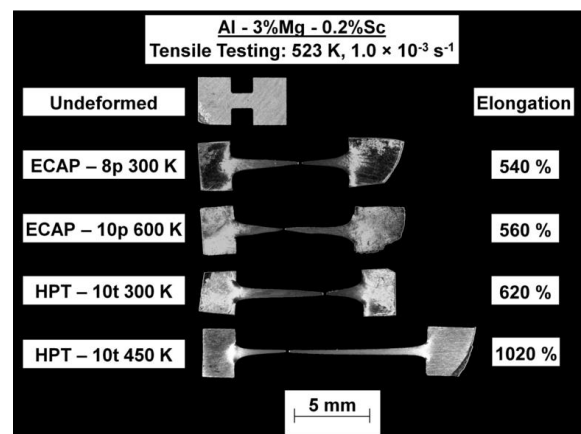


Fig. 3 Examples of superplasticity in an Al-3% Mg-0.2% Sc alloy after processing by ECAP for 8 or 10 passes or HPT for 10 turns (t) [21].

## An examination of superplasticity in Al-Mg-Sc alloys

Figure 4 shows an example of the remarkable superplastic elongations that may be achieved in an Al-3% Mg-0.2% Sc alloy after processing by ECAP for 8 passes at room temperature using a die with an internal angle of  $90^\circ$  [23]. These elongations are up to  $>2000\%$  at high strain rates in the range of  $10^{-3}$  to  $10^{-1} \text{ s}^{-1}$ . By contrast, processing by cold rolling (CR) produces a microstructure that is similar in appearance but the boundaries then have low angles of misorientation so that GBS is not a potential flow mechanism and the material in this condition is not superplastic.

There are numerous procedures for achieving small grains sizes in Al-Mg-Sc alloys and Figs 5-8 summarize experimental data from numerous reports where the references associated with these diagrams were given earlier [21].

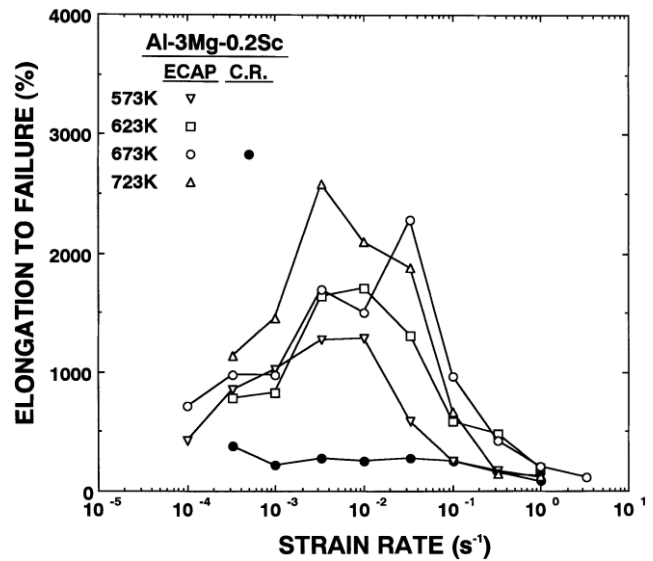


Fig. 4 Examples of excellent superplasticity in an Al-3% Mg-0.2% Sc alloy after processing by ECAP but with an absence of superplasticity after processing by cold rolling (CR) [23].

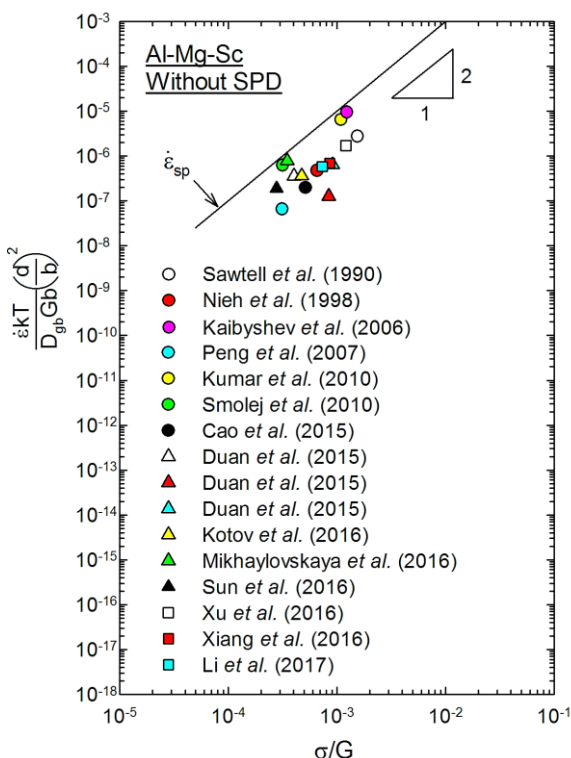


Fig. 5 Temperature and grain size compensated strain rate versus normalized stress for superplastic Al-Mg-Sc alloys without SPD processing [21]

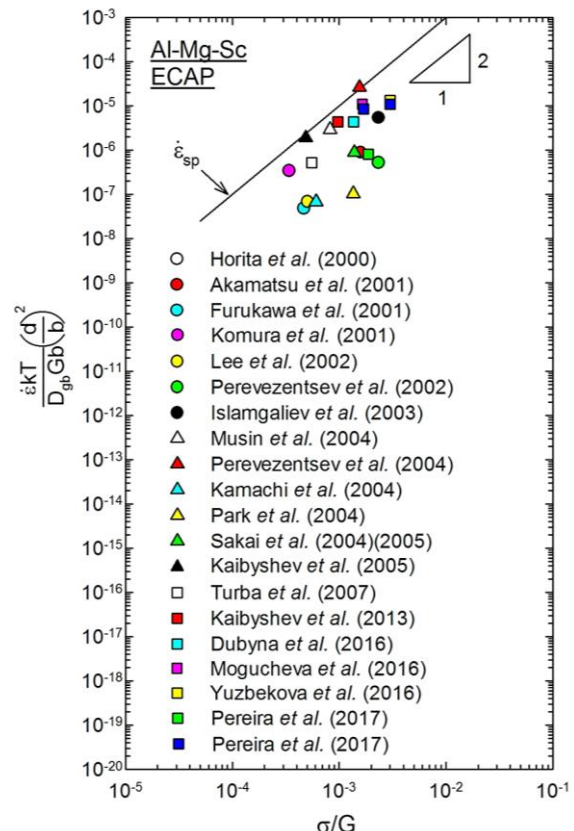


Fig. 6 Temperature and grain size compensated strain rate versus normalized stress for superplastic Al-Mg-Sc alloys processed by ECAP [21].

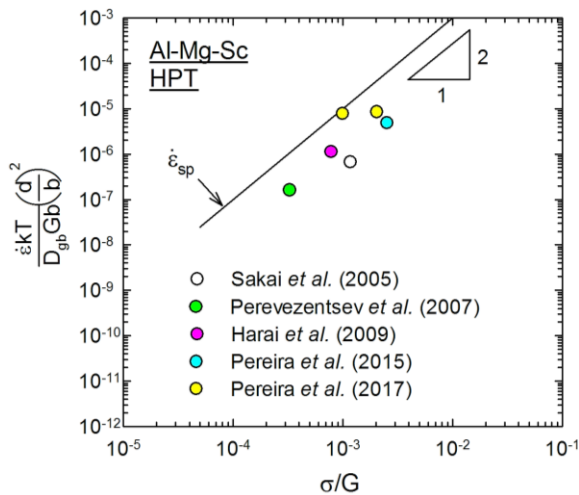


Fig. 7 Temperature and grain size compensated strain rate versus normalized stress for superplastic Al-Mg-Sc alloys processed by HPT [21].

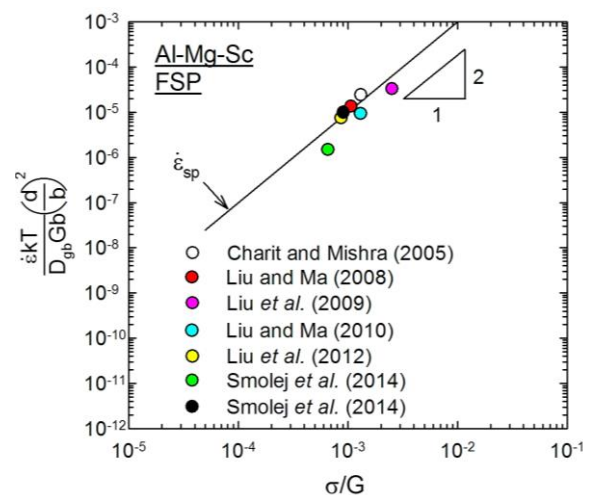


Fig. 8 Temperature and grain size compensated strain rate versus normalized stress for superplastic Al-Mg-Sc alloys processed by FSP [21].

In Fig. 5, results are shown for samples not subjected to any SPD processing where the small grains were obtained using conventional thermo-mechanical processing. These experimental points are taken from various reports and the upper solid line is the theoretical prediction using Eq. (1) for conventional superplastic flow. Similar plots are given in Fig. 6 for samples processed by ECAP, in Fig. 7 for samples processed by HPT and in Fig. 8 for samples obtained after friction stir processing (FSP) where this is a solid-state joining technique that produces an ultrafine-grained microstructure over a limited area. For each plot, the upper solid line again denotes the prediction using Eq. (1). Inspection shows that all of these results are mutually consistent and all datum points lie close together within one order of magnitude of strain rate. Furthermore, all results are in reasonable consistency with the predictions of Eq. (1). This confirms the value of using SPD processing to attain the very small grain sizes that can be used in developing and exploiting superplastic properties in the Al-Mg-Sc alloys [24,25]. For convenience, complete tabulations of all experimental data for superplastic Al-Mg-Sc alloys were given earlier [21].

## Summary and conclusions

1. Al-Mg-Sc alloys are excellent candidate materials for achieving high superplastic elongations when pulling in tension. Experimental results show elongations of more than 2000% at relatively fast strain rates.
2. An analysis of published data for a large number of Al-Mg-Sc alloys shows that the results are mutually consistent and in reasonable agreement with the theoretical model for superplastic flow.

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## References

- [1] T.G. Langdon, Seventy-five years of superplasticity: historic developments and new opportunities, *J. Mater. Sci.* 44 (2009) 5998-6010.

- [2] G.D. Bengough, A study of the properties of alloys at high temperatures, *J. Inst. Metals* 7 (1912) 123-178.
- [3] C.H.M. Jenkins, Strength of Cd-Zn and Sn-Pb alloy solder, *J. Inst. Metals* 40 (1928) 21-32.
- [4] C.E. Pearson, The viscous properties of extruded eutectic alloys of lead-tin and bismuth-tin, *J. Inst. Metals* 54 (1934) 111-124.
- [5] A.A. Presnyakov, *Sverkhplastichnost' Metallov I Splavov*, Nauka, Alma-Ata, U.S.S.R., 1969. (English translation: C.B. Marinkov, *Superplasticity of Metals and Alloys*, The British Library. Wetherby U.K., 1976).
- [6] E.E. Underwood, A review of superplasticity and related phenomena, *JOM* 14 (1962) 914-919.
- [7] W.A. Backofen, L.R. Turner, D.H. Avery, Superplasticity in an Al-Zn alloy, *Trans. ASM* 57 (1964) 980-990.
- [8] R. Grimes, Superplastic forming: evolution from metallurgical curiosity to major manufacturing tool? *Mater. Sci. Tech.* 19 (2003) 3-10.
- [9] A.J. Barnes, Superplastic forming 40 years and still growing, *J. Mater. Eng. Perform.* 16 (2007) 440-454.
- [10] T.G. Langdon, The mechanical properties of superplastic materials, *Metall. Trans. A* 13A (1982) 689-701.
- [11] T.G. Langdon, An evaluation of the strain contributed by grain boundary sliding in superplasticity, *Mater. Sci. Eng. A* 174 (1994) 225-230.
- [12] L.K.L. Falk, P.R. Howell, G.L. Dunlop, T.G. Langdon, The role of matrix dislocations in the superplastic deformation of a copper alloy, *Acta Metall.* 34 (1986) 1203-1214.
- [13] R.Z. Valiev, T.G. Langdon, An investigation of the role of intragranular dislocation strain in the superplastic Pb-62% Sn eutectic alloy, *Acta Metall. Mater.* 41 (1993) 949-954.
- [14] Y. Xun, F.A. Mohamed, Slip-accommodated superplastic flow in Zn-22% Al, *Philos. Mag.* 83 (2003) 2247-2266.
- [15] Y. Xun, F.A. Mohamed, Superplastic behavior of Zn-22% Al containing nanoscale dispersion particles, *Acta Mater.* 52 (2004) 4401-4412.
- [16] F.A. Mohamed, T.G. Langdon, Deformation mechanism maps for superplastic materials, *Scripta Mater.* 10 (1976) 759-762.
- [17] T.G. Langdon, A unified approach to grain boundary sliding in creep and superplasticity, *Acta Metall. Mater.* 42 (1994) 2437-2443.
- [18] M. Kawasaki, T.G. Langdon, Review: achieving superplastic properties in ultrafine-grained materials at high temperatures, *J. Mater. Sci.* 51 (2016) 19-32.
- [19] S. Lee, A. Utsunomiya, H. Akamatsu, K. Neishi, M. Furukawa, Z. Horita, T.G. Langdon, Influence of scandium and zirconium on grain stability and superplastic ductilities in ultrafine-grained Al-Mg alloys, *Acta Mater.* 50 (2002) 553-564.
- [20] J. Wang, Z. Horita, M. Furukawa, M. Nemoto, N.K. Tsenev, R.Z. Valiev, Y. Ma, T.G. Langdon, An investigation of ductility and microstructural evolution in an Al-3% Mg alloy with submicron grain size, *J. Mater. Res.* 8 (1993) 2810-2818.
- [21] P.H.R. Pereira, Y. Huang, M. Kawasaki, T.G. Langdon, An examination of the superplastic characteristics of Al-Mg-Sc alloys after processing, *J. Mater. Res.* 32 (2017) 4541-4553.
- [22] T.G. Langdon, Fracture processes in superplastic flow, *Metal Sci.* 16 (1982) 175-183.
- [23] S. Komura, Z. Horita, M. Furukawa, M. Nemoto, T.G. Langdon, An evaluation of the flow behavior during high strain rate superplasticity in an Al-Mg-Sc alloy, *Metall. Mater. Trans. A* 32A (2001) 707-716.
- [24] T.G. Langdon, Twenty-five years of ultrafine-grained materials: Achieving exceptional properties through grain refinement, *Acta Mater.* 61 (2013) 7035-7059.
- [25] M. Kawasaki, T.G. Langdon, The contribution of severe plastic deformation to research on superplasticity, *Mater. Trans.* 60 (2019) 1123-1130.